

FORM PTO-1390
(REV 11-98)

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER

**TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371**

LC-377-PCT-US

U.S. APPLICATION NO. (if known, see 37 CFR 1.5)

10/018808

INTERNATIONAL APPLICATION NO.
PCT/US00/20076INTERNATIONAL FILING DATE
21 JULY 2000 (21.07.00)PRIORITY DATE CLAIMED
21 JULY 1999 (21.07.99)

TITLE OF INVENTION

Washable Impregnation Compositions

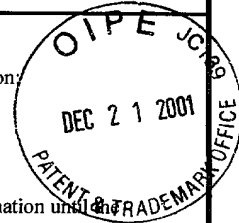
APPLICANT(S) FOR DO/EO/US NEWBERTH, Frederick F. III, and CHUPAS, Peter J.

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(l).
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
 - a. ☒ is transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☒ has been transmitted by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☐ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☒ A copy of the International Search Report (PCT/ISA/210).
8. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
 - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ have been transmitted by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☐ have not been made and will not be made.
9. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
10. ☒ An executed declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
11. ☒ A copy of the International Preliminary Examination Report (PCT/IPEA/409).
12. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 13. to 19. below concern document(s) or information included:

13. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98, listing the documents cited in the International Search Report and enclosing copies thereof.
14. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
15. ☐ A **FIRST** preliminary amendment.
☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
16. ☐ A substitute specification.
17. ☐ A change of power of attorney and/or address letter.
18. ☐ Certificate of Mailing by Express Mail.
19. ☒ Other items or information: Demand, Written Opinion and Response thereto and a RETURN RECEIPT POSTCARD



U.S. APPLICATION NO (If known, see 37 CFR 1.5.) 10/018808		INTERNATIONAL APPLICATION NO PCT/US00/20076		ATTORNEY'S DOCKET NUMBER LC-377-PCT-US	
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20. <input checked="" type="checkbox"/> The following fees are submitted: BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)): Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO\$1,000.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO\$860.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO\$710.00 International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4)\$690.00 International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4)\$100.00 ENTER APPROPRIATE BASIC FEE AMOUNT = \$100.00				CALCULATIONS PTO USE ONLY	
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input checked="" type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)). + \$					
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	20 - 20 =	0	X \$ 18.00	\$ - 0 -	
Independent claims	4 - 3 =	1	X \$ 80.00	\$ 80.00	
MULTIPLE DEPENDENT CLAIM(S) (if applicable) + \$270.00				\$ - 0 -	
TOTAL OF ABOVE CALCULATIONS =				\$180.00	
Reduction of 1/2 for filing by small entity, if applicable. A Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28).				\$ ---	
SUBTOTAL =				\$180.00	
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)). + \$ ---				\$ ---	
TOTAL NATIONAL FEE =				\$180.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property + \$ ---				\$ ---	
TOTAL FEES ENCLOSED =				\$0.00	
				Amount to be:	
				Refunded	\$
				charged	\$

a. ☐ A check in the amount of \$ _____ to cover the above fees is enclosed.

b. ☒ Please charge my Deposit Account No. **12-2135** in the amount of \$ **180.00** to cover the above fees. A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to my Deposit Account No. **12-2135**. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

STEVEN C. BAUMAN
LOCTITE CORPORATION
1001 TROUT BROOK CROSSING
ROCKY HILL, CT 06067 USA

SIGNATURE _____

NAME **STEVEN C. BAUMAN**

REGISTRATION NUMBER **33,832**

500-82 PCT

PATENT

WASHABLE IMPREGNATION COMPOSITIONSFIELD OF THE INVENTION:

The present invention relates generally to washable, polymerizable compositions. More particularly, the present invention relates to such compositions curable through mechanisms, anaerobic, and heat curing for use as impregnation sealants.

5 BRIEF DESCRIPTION OF RELATED TECHNOLOGY:

Impregnation sealing of porosity in porous parts frequently is carried out by introducing sealant compositions into the porosity under a pressure differential, by vacuum techniques which are well known in the art.

10 Sealant compositions typically employed in these impregnation applications include a wide variety of self-curing anaerobic sealants, e.g., the compositions described in U.S. Patent Nos. 3,672,942; 3,969,552; Re. 32,240; and 4,632,945, which are curable through free-radical polymerization in the presence of suitable free-radical initiators, e.g., peroxy-type initiators, as well as thermal-curing sealants, e.g., the compositions described in U.S. Patent Nos.
15 4,416,921 and 4,416,921, as well as sealants which cure by both anaerobic and heat cure mechanisms.

One problem common to many impregnation sealants is the accumulation of excess sealant on the outer surface of parts. Excess sealant is removable by normal abrasion or by
20 contact with various liquids. The removal of extraneous or surface accumulation of anaerobic and heat curing sealants from the parts is important because such residues can readily contaminant the environment of porous parts. In addition, such surface sealant deposits may, by virtue of their thickness, cause the impregnated product part to vary from the desired dimensional specifications. This often renders the part deficient or even useless for its
25 intended function in applications requiring close dimensional tolerances.

Furthermore, such surface sealant deposits may interfere with subsequent painting,

plating, or assembly operations or cause delamination of applied paint or plated films which frequently are performed on porous articles subsequent to their impregnation. Specifically, such surface sealant deposits may be removed during painting or plating operations, resulting in contamination of the baths used in such operations, and may interfere with the adhesion of paint, plating, and the like to the impregnated part.

To remove excess sealant from impregnated articles, agitated rinse times of significant duration are required. The actual rinse time will depend upon, among other things, the nature of the article, such as porosity, and the washability of the uncured sealant in an aqueous solution. Often such rinse operations are from about five to about twenty minutes, but actual rinse times may for any particular article may be even longer in duration. In addition, chemicals, such as surfactants or detergents, may also be added to the aqueous solution to facilitate the removal of sealant deposits.

For example, U.S. Patent No. 3,672,942 to Neumann et al. discloses an anaerobic impregnant comprising a free-radical polymerizable acrylate ester monomer and free-radical polymerization initiator, which requires an organic solvent, such as a halogenated hydrocarbon, to remove uncured impregnant from the outer surface of a porous article.

U.S. Patent No. 3,969,552 to Malofsky et al. describes a washing process for removing excess impregnant from the surface of the porous article after porosity impregnation. The disclosed impregnation composition comprises an acrylic anaerobic curing resin and a peroxy initiator therefor. The wash solution is an aqueous solution of a nonionic surfactant of specified formula which is necessary for the removal of uncured impregnant.

U.S. Patent No. Re. 32,240 to DeMarco describes a self-emulsifying anaerobic composition for porosity impregnation applications, comprising an anaerobically curing monomer such as an acrylic ester, a peroxy initiator therefor, e.g., a hydroperoxide or perester, an anionic or nonionic surfactant which is dissolved in the composition and renders

it self-emulsifying upon mixing with water.

U.S. Patent No. 5,256,450 to Catena describes an anaerobic polymerizable acrylate composition which requires a mixture of three different polymerizable acrylates in specific amounts to obtain a composition that cures and rinses without the use of organic solvents or surfactants.

The above-described anaerobic sealant compositions are typically impregnated into the porosity of metal parts by vacuum and pressure techniques. A vacuum removes air from the porosity of the metal parts. Sealant compositions are then introduced into the porosity under a pressure differential using ambient pressure or elevated pressure conditions. After impregnation, an operation, such as a centrifuge operation, removes excess surface sealant from the metal part. Even after such removal of gross surface accumulations of the impregnant, there is a significant amount of impregnant at the surface of the porous articles, particularly in the vicinity of the pores. When the impregnant is anaerobically cured, the aforementioned surface accumulations as well as the outermost layer of the impregnant in the pores of the article, particularly shallow surface pores, are in contact with oxygen, so that such surface quantities of the impregnant are uncured or only partially cured.

Remaining surface sealant or sealant trapped in blind holes of the impregnated parts is typically removed in an agitated water rinse zone. The impregnated and water-rinsed parts may be transferred to an activator zone in which the impregnated parts are contacted with a catalyst activator solution, to effect curing of the sealant material at the entrance to the pores in the parts. This creates a hardened plug or cap of sealant material in the outer portion of the pore, trapping the resin for anaerobic self-cure.

Thereafter, the impregnated parts may be transferred to a final rinse zone for removal of the activator solution from the impregnated parts. This final rinse solution may be at elevated temperature, e.g., on the order of about 50°C, to warm the impregnated parts for quick drying, and to accelerate curing of the anaerobic impregnant within the interior porosity

of the article, the rate of such cure increasing with increasing temperature.

As a variation on the above-described impregnation system, it is known to utilize a heat-curing resin in place of the anaerobically-curing resin, whereby the activating and final rinsing steps previously described are eliminated in favor of a hot rinse final step. In the heat-curing resin impregnation system, after impregnation and rinsing of excess surface material, the parts are contacted with hot water at temperatures on the order of about 50°C to about 90°C to cure the impregnant resin.

Among the previously developed heat-curing impregnating compositions for sealing porous parts are the compositions disclosed in the patents identified and discussed below.

U.S. Patent No. 4,416,921 to Dunn describes a heat-curing sealant composition which contains a polymerizable acrylic monomer, an azonitrile and a anionic or nonionic surfactant to render the composition self-emulsifying upon mixing with water.

U.S. Patent No. 4,147,821 to Young describes a heat-curing sealant composition which contains (meth)acrylic monomer and a polyfunctional acrylic monomer. An emulsifier is required to aid in the rinsing of uncured sealant from the surface of a porous article.

Once the heat-curable impregnant composition is introduced into the porosity of the parts to be sealed, the parts are transferred to an agitated water rinse zone for removal of any remaining surface accumulations of sealant or extraneous sealant which is trapped in blind holes of the impregnated parts. After removal of the excess sealant in the agitated water rinse zone, the impregnated parts are passed to a tank containing hot water, e.g., at a temperature of 90°C to 150°C, or other medium at elevated temperature which serves to cure the sealant composition in the porosity. Relative to anaerobic impregnant compositions, heat-curable impregnant compositions may be effectively used with a minimum of monitoring and maintenance, with little or no aeration being required.

In all of the above-described impregnation compositions and systems, either organic solvents or specific surfactants are used to remove uncured sealant in a reasonable rinse time or specific multi-component sealant compositions are used to avoid excessive rinse times.

5 Accordingly, there is a need to provide a heat-curable and/or an anaerobic impregnating sealant without these and other disadvantages.

SUMMARY OF THE INVENTION:

10 The present invention provides washable compositions for sealing porous articles which have improved washability characteristics and reduced rinsing requirements. The present compositions achieve lower rinse times while producing improved surface cleaning of uncured polymer. The compositions of the present invention demonstrate utility in the sealing and/or aqueous rinsing operations, and obviate the conventional use of multi-component cleaning systems.

15 In particular, the present invention provides a sealant composition with improved washability, thereby reducing the rinse duration, improved ease of use by eliminating the need for specific surfactants, and which improve surface cleanliness of the porous article.

20 In one embodiment of the present invention, the inventive composition includes a curable (meth)acrylate glycerol, and is self-emulsifying upon mixing with water to facilitate aqueous rinsing of uncured composition. The inventive composition further includes curing initiators and curing accelerators to promote anaerobic or thermal curing through free radical mechanisms.

25 In another embodiment of the present invention, the invention composition includes a polymerizable composition and further includes a compound selected from the group consisting of glycerol, oxylated glycerol, (meth)acrylate glycerol and combinations thereof which improve the washability of the inventive sealants in aqueous solutions.

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In one desirable embodiment, the inventive composition contains an (meth)acrylate glycerol which has at least one terminal (meth)acrylate group to allow crosslinking of the (meth)acrylate glycerols upon curing.

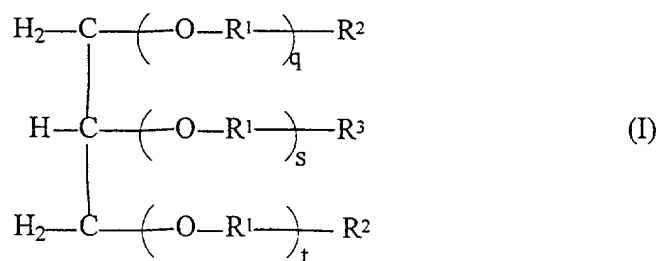
5 **DETAILED DESCRIPTION OF THE INVENTION:**

10 The present invention is directed to a sealant composition with improved washability characteristics as compared to known sealant compositions. The present invention provides sealant compositions, the components of which serve to provide washability and self-emulsificability to the overall composition. These components may be included with the inventive sealant composition during the impregnation operation or may be incorporated during the water rinse operation to provide washability and self-emulsificability to the overall composition. Uncured sealant is typically removed from the surface of the porous article during the aqueous rinse portion of the sealing process. As used herein the term "washable" and its variants refer to the ability of a sealant composition to emulsify in an aqueous solution and be readily removed from unwanted areas of an article in the aqueous solution. Also, as used herein the term "self-emulsificability" and its variants refer to the ability of one liquid to form minute droplets in a second liquid resulting in a heterogeneous mixture of two liquid phases.

20 The present compositions employ an independent component selected from the group consisting of glycerol, oxylated glycerol, (meth)acrylate glycerol and combinations thereof as an additive to anaerobic or heat curable polymerizable compositions. The polymerizable composition includes polyfunctional and monofunctional (meth)acrylate esters to effectuate the polymerizable properties of the sealant. The inventive sealant compositions may contain other components to tailor the polymerizing, curing or emulsifying properties of the compositions. The inventive sealants also contain an initiator system and/or inhibitor systems to provide controlled anaerobic or thermal curing mechanisms. These compositions have a variety of uses, including an impregnation compositions, sealants, adhesives, coatings and the like. One particularly desirable embodiment relates to impregnation sealant compositions for porous parts.

The inventive compositions are generally curable by free-radical mechanisms. Typically, anaerobic conditions or elevated temperature condition may be used. In impregnation applications, however, generally anaerobic and/or thermal mechanisms are used. Furthermore, the inventive compositions are self-emulsifying upon mixing with water to facilitate the aqueous rinsing of an impregnated article.

In one embodiment of the inventive composition the washability and self-emulsificability enhancing component in the form of glycerol is incorporated into the resultant polymer backbone. This composition includes a curable (meth)acrylate glycerol component having the formula:



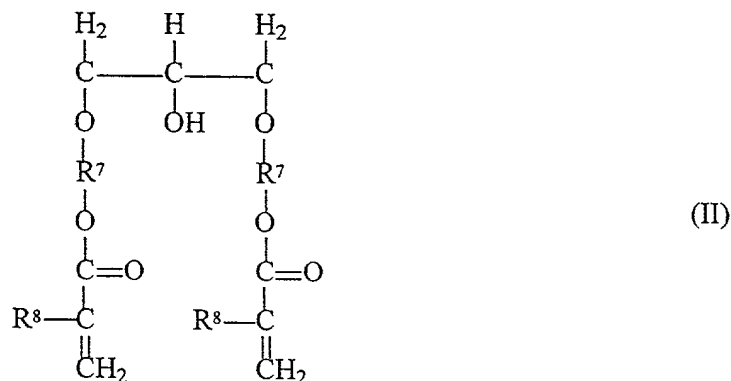
wherein R¹ is a substituted or unsubstituted C₁ to C₅ alkyl or a combination thereof, R² and R³ are independently selected from the group consisting of hydroxyl, (meth)acrylate and combinations thereof, provided that at least one R² is a (meth)acrylate, where q, s and t are independently from about 0 to about 35; and a free radical initiator component or system for producing free radicals to initiate cure of said composition. The short chain C₁ to C₅ alkyl groups, as compared to longer chain alkyl groups, assist in the washability of the inventive compositions. Longer chain alkyl groups can act negatively to retard emulsification. Desirably, R¹ is an ethyl, a propyl or combinations thereof. The (meth)acrylate glycerol compositions produce cross-linked polymers upon curing which serve as effective and durable self-washing sealants. The free radical initiator component provides free radicals for both anaerobic or heat curing of the composition.

The inventive compositions may contain from about 50% to about 99%

(meth)acrylate glycerol by weight of the total composition with the balance including other materials, for instance, initiators, inhibitors, surfactants, inerts, for instance, non-reactive plasticizers, and the like.

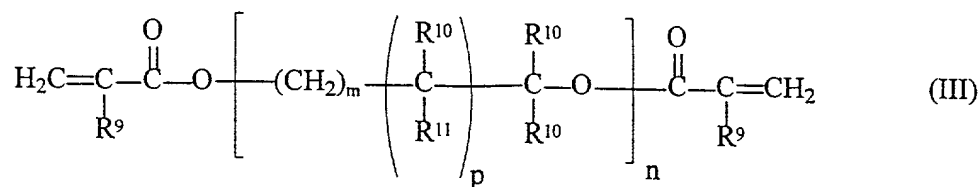
- 5 The (meth)acrylate glycerol compositions may be suitably prepared by condensing hydroxyalkyl (meth)acrylate, such as hydroxyethyl(meth)acrylate (HEMA), onto hydroxyl groups of an oxylated glycerol, such as an oxylated glycerol of formula VI below.

- 10 In an alternate embodiment, the inventive compositions include a self-washing polymerizable di(meth)acrylate glycerol having the formula:

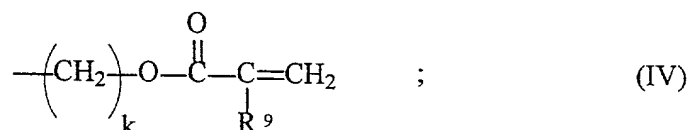


wherein R⁷ is an ethyl or propyl alkyl and R⁸ is hydrogen or methyl; and a free radical initiator component or system.

- 15 Furthermore, in still another embodiment the inventive sealant composition may include at least one crosslinkable polymer, a curing component for said polymer and a compound selected from the group consisting of glycerol, oxylated glycerol, (meth)acrylate glycerol and combinations thereof. Desirably, the polymerizable component has a majority of polyfunctional (meth)acrylate esters (hereinafter, poly(meth)acrylate esters). These
- 20 polyfunctional esters produce cross-linked polymers, which serve as effective and durable sealants, adhesives and coatings. While various (meth)acrylate esters may be used, desirable poly(meth)acrylate esters include those with the following general formula:

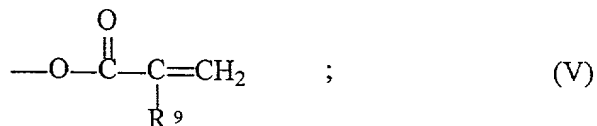


wherein R^{10} represents a radical selected from the group consisting of hydrogen, lower alkyl of from 1 to about 4 carbon atoms, hydroxyalkyl of from 1 to about 4 carbon atoms and



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R^9 is a radical selected from the group consisting of hydrogen, halogen, and lower alkyl of from 1 to about 4 carbon atoms; R^{11} is a radical selected from the group consisting of hydrogen, hydroxyl, and



10 and m may be 0 to 12, and desirably from 0 to about 6; n is equal to at least 1, e.g., 1 to about 20 or more, and desirably between about 2 to about 6; k is 1 to about 4; and p is 0 or 1.

The polymerizable poly(meth)acrylate esters corresponding to the above general formula are exemplified by, but not restricted to, the following materials: di-, tri- and
 15 tetraethyleneglycol dimethacrylate, dipropyleneglycol dimethacrylate; polyethyleneglycol dimethylacrylate (PEGMA); di(pentamethyleneglycol) dimethacrylate; tetraethyleneglycol diacrylate; tetraethyleneglycol di(chloracrylate); diglycerol diacrylate; diglycerol tetramethacrylate; tetramethylene dimethacrylate; ethylene dimethacrylate; and
 20 neopentylglycol diacrylate. Combinations and derivatives of these polyfunctional materials are contemplated.

Monofunctional (meth)acrylate esters (esters containing one (meth)acrylate group) are also advantageously used in the present compositions. The most common of these monofunctional esters include the alkyl esters such as lauryl methacrylate. Many of the lower molecular weight alkyl esters are quite volatile, and frequently it is more desirable to use a higher molecular weight homolog, such as decyl methacrylate or dodecyl methacrylate, or any other fatty acid acrylate esters, in (meth)acrylate-based impregnant compositions.

When monofunctional (meth)acrylate esters are employed in the present compositions, it is desirable to use an ester which has a relatively polar alcohol moiety. Such materials are less volatile than low molecular weight alkyl esters and, in addition, the polar group tends to provide intermolecular attraction in the cured polymer, thus producing a more durable seal. Desirably the polar group is selected from the group consisting of labile hydrogen, heterocyclic ring, hydroxy, amino, cyano, and halogen polar groups. Typical examples of compounds within this category are cyclohexylmethacrylate, tetrahydrofurfuryl methacrylate, hydroxyethyl acrylate (HEMA), hydroxypropyl methacrylate (HPMA), t-butylaminoethyl methacrylate, cyanoethylacrylate, and chloroethylmethacrylate. Combinations of monofunctional (meth)acrylate are contemplated.

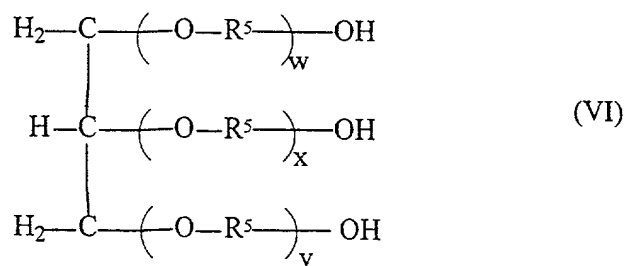
When poly(meth)acrylate esters and monofunctional (meth)acrylate esters are employed together in the present compositions, the ratio of poly(meth)acrylate esters to monofunctional (meth)acrylate esters on a weight basis is generally about 0.05:1 to about 20:1. Desirably, for use in impregnation compositions the ratio is about 5:1. Furthermore, the inventive curable (meth)acrylate glycerol compounds can be combined with such esters in amounts of about 0.1% to about 99% by weight of the total composition. In other words, the washability of such esters can be improved by combining various amounts of (meth)acrylate glycerol.

In yet another embodiment of the present invention, a glycerol or an oxylated glycerol is combined with polymerizable sealant compositions to further improve washability thereof.

The oxylated glycerol may be incorporated into the sealant composition or may be added

separately into the aqueous rinse solution used to wash the sealant compositions thereby aiding in the removal of uncured surface and extraneous sealant from the surface of the part.

The oxylated glycerols of the present invention have the formula:



wherein R^s is a substituted or unsubstituted C_1 to C_5 alkyl or a combination thereof and w , x and y are independently from about 0 to about 35. Desirably, R^s is an ethyl, a propyl or combinations thereof.

When poly(meth)acrylate esters and monofunctional (meth)acrylate esters are employed together in the present compositions, the ratio of poly(meth)acrylate esters to monofunctional (meth)acrylate esters on a weight basis is generally about 0.05:1 to about 20:1. Desirably, for use in impregnation compositions the ratio is about 5:1. Furthermore, the inventive oxylated glycerols can be combined with such esters in amounts of about 0.1% to about 75% by weight of the total composition. In other words, the washability of such esters can be improved by combining various amounts of oxylated glycerols.

The compositions of the present invention may be anaerobically curable through a free-radical mechanism, with an initiator being present therein, or an initiator system comprising a redox polymerization initiator (i.e., an ingredient or a combination of ingredients which produce an oxidation-reduction reaction, resulting in the production of free radicals). Suitable initiators include peroxy materials e.g., peroxides, hydroperoxides, and peresters, which are capable of inducing polymerization of the inventive compositions in the substantial absence of oxygen, and yet not induce polymerization as long as oxygen is present. Organic hydroperoxides are the desirable peroxy materials with t-butyl

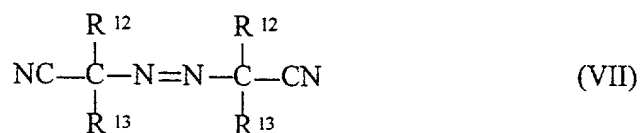
hydroperoxide and cumene hydroperoxide being particularly useful with the inventive compositions.

In addition to initiator components, the composition of the present invention may include various initiator accelerators, as for example hydroperoxide decomposition accelerators, when hydroperoxides are used as cure initiators in the sealant material. Typical examples of potentially suitable accelerators include: tertiary amines such as tributyl amine; sulfimides such as benzoic sulfimide (or saccharin); formamide; and compounds containing transition metals, such as copper octanoate.

The inventive compositions may also be heat-curable compositions through a free-radical mechanism, with a heat-cure initiator being present therein, or an initiator system comprising a redox polymerization initiator (i.e., an ingredient or a combination of ingredients which at the desired elevated temperature conditions, e.g. from about 90° to about 150°C, produce an oxidation-reduction reaction, resulting in the production of free radicals). Suitable initiators may include peroxy materials, e.g., peroxides, hydroperoxides, and peresters, which under appropriate elevated temperature conditions decompose to form peroxy free radicals which are initiatingly effective for the polymerization of the inventive compositions.

Another useful class of heat-curing initiators comprises azonitrile compounds which yield free radicals when decomposed by heat. Heat is applied to cure the composition, and the resulting free radicals initiate polymerization of the inventive composition.

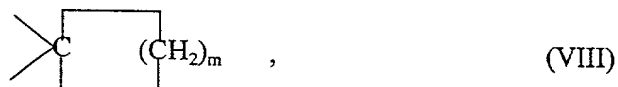
For example, azonitrile may be a compound of the formula:



wherein R¹² is a methyl, ethyl, n-propyl, iso-propyl, iso-butyl or n-pentyl radical, and R¹³ is a

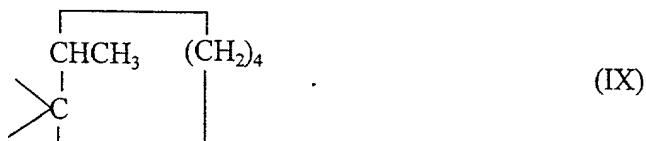
methyl, ethyl, n-propyl, iso-propyl, cyclopropyl, carboxy-n-propyl, iso-butyl, cyclobutyl, n-pentyl, neo-pentyl, cyclopentyl, cyclohexyl, phenyl, benzyl, p-chlorbenzyl, or p-nitrobenzyl radical or R¹² and R¹³, taken together with the carbon atom to which they are attached, represent a radical of the formula

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wherein m is an integer from 3 to 9, or the radical

10



Compounds of the above formula are more fully described in U.S. Patent No. 4,416,921, the disclosure of which hereby is incorporated herein by reference.

15

Azonitrile initiators of the above-described formula are readily commercially available, e.g., the initiators which are commercially available under the trademark VAZO® from E. I. DuPont de Nemours and Company, Inc. (Wilmington, Del.), including VAZO® 52 (R¹²=methyl, R¹³=isobutyl), VAZO® 64 (R¹²=methyl, R¹³=methyl), and VAZO® 67 (R¹²=methyl, R¹³=ethyl), all such R¹² and R¹³ constituents being identified with reference to the

20

above-described azonitrile general formula.

A desirable azonitrile initiator is 2,2'-azobis(iso-butyronitrile) or AZBN.

25

The azonitrile may be employed in the inventive heat-curable compositions in concentrations on the order of about 500 to about 10,000 parts per million (ppm) by weight, desirably about 1000 to about 5000 ppm.

30

Other (meth)acrylic monomer-based impregnant compositions of a heat-curable character may be employed in the broad practice of the present invention, including those disclosed in UK Patent Specifications 1,308,947 and 1,547,801. As described in these

references, the monomeric impregnant composition may contain suitable inhibitors serving to restrict or preclude the occurrence of polymerization of the monomer, at temperatures below those desired or recommended for heat-curing of the impregnant composition.

5 The inventive impregnant compositions may also contain other constituents, such as:
other co-monomer species, reactive diluents, pigments, surfactants, fillers, polymerization
inhibitors, stabilizers, anti-oxidants, anti-corrosion additives, and the like. For example,
surfactants may be combined with the inventive compositions or included in the aqueous
rinse solution. The use of surfactants and specific materials utilized for such purpose are
10 more fully described in U.S. Patent No. 3,969,552 and Re. 32,240, the disclosures of each of
which are expressly incorporated herein by reference. Suitable surfactants include classes of
anionic surfactants, such as petroleum sulfonates, alkyl sulfonates or alkylaryl sulfonates and
nonionic surfactants, such as, ethoxylated alkyl phenols, ethoxylated linear secondary
alcohols, polyoxyethylene or polyoxypropylene glycols.

15 The invention may be further understood with reference to the following non-limiting
examples. Percent weights are per the total composition unless otherwise specified.

EXAMPLES

Example 1

- 5 An anaerobic sealant composition according to the present invention (Composition One) was prepared with the following formulation:

Table 1

<u>Composition One</u>	<u>WT %</u>
Triethyleneglycol dimethacrylate	74.00
Lauryl methacrylate	15.00
Hydroxpropyl methacrylate	5.00
Surfactant	5.24
Fluorescence	0.02
Inhibitor	0.04
Benzosulfimide (saccharin)	0.30
t-butyl hydroperoxide	0.40
Total:	100.00

- 10 A lapshear, such as a metal lapshear (1" x 4" x 1/16") in accordance with ASTM D-1002, was coated with the above inventive composition. The coated lapshear was repeatedly dipped into room temperature tap water to clean the coated lapshear to yield a base dipping requirement to clean the lapshear. Glycerol dimethacrylate was then combined with inventive Composition One at various levels as shown below and the cleaning procedure was repeated.
- 15 As illustrated in the below results in Table 2, including glycerol dimethacrylate in the Composition One increased the washability thereof, as evidenced in a decrease in dips required for cleaning. These compositions proved to have particular efficacy as an impregnation composition.

Table 2

<u>Glycerol Dimethacrylate Incorporated into Inventive Composition One, WT%</u>	<u>Number of Dips into 27°C Tap Water to Clean Coated Lapshear</u>
0	50+
1	35
3	35
5	35
7	25
9	20
15	20

Example 2

5

An anaerobic sealant composition according to the present invention (Composition Two) was prepared with the following formulation:

Table 3

<u>Composition Two</u>	<u>WT %</u>
Butanediol dimethacrylate	50.00
Triallyl Cyanurate	30.00
Lauryl methacrylate	19.04
Fluorescence	0.02
Inhibitor	0.04
Saccharin	0.30
70% t-butyl hydroperoxide	0.60
Total:	100.00

A lapshear, as described above in Example 1, was coated with the above composition.

- 10 The coated lapshear was repeatedly dipped into room temperature tap water to clean the coated lapshear to yield a base dipping requirement to clean the lapshear. Glycerol dimethacrylate was then combined with inventive Composition Two at various levels as shown below and the cleaning procedure was repeated. As illustrated in the results below, incorporating glycerol dimethacrylate in the composition increased the washability thereof, as
- 15 evidenced by the decrease in number of dips required for cleaning. These compositions

proved to have particular efficacy as an impregnation composition.

Table 4

<u>Glycerol Dimethacrylate Incorporated into Inventive Composition Two, WT%</u>	<u>Number of Dips into 27°C Tap Water to Clean Coated Lapshear</u>
0	50+
1	15
3	13
5	10
7	8
9	8

Example 3

5

The anaerobic sealant composition (Composition Two) of Example 2 was used in the following washability study.

A lapshear, as described above in Example 1, was coated with the above composition.

10 The coated lapshear was repeatedly dipped into room temperature tap water to clean the coated lapshear to yield a base dipping requirement to clean the lapshear. Ethoxylated glycerol was then combined with the sealant composition at various levels as shown below and the cleaning procedure was repeated. As illustrated in the below results in Table 5, including ethoxylated glycerol in the composition increased the washability thereof, as
15 evidenced by a decrease in the number of dips required for cleaning. These compositions proved to have particular efficacy as an impregnation composition.

Table 5

<u>Ethoxylated Glycerol Incorporated into Inventive Composition Two, WT%</u>	<u>Number of Dips into 27°C Tap Water to Clean Coated Lapshear</u>
0	50+
1	40
3	20
5	13
7	13
9	10

Example 4

The anaerobic sealant composition (Composition Two) of Example 2 was used in the following washability study.

A lapshear, as described above in Example 1, was coated with the above sealant composition. The coated lapshear was repeatedly dipped into room temperature tap water to clean the coated lapshear to yield a base dipping requirement to clean the lapshear. Propoxylated glycerol was then combined with the sealant composition at various levels as shown below and the cleaning procedure was repeated. As illustrated in the below results in Table 6, including propoxylated glycerol in the sealant composition increased the washability thereof, as evidenced by a decrease in the number of dips required for cleaning. These compositions proved to have particular efficacy as an impregnation composition.

Table 6

<u>Propoxylated Glycerol Incorporated into Inventive Composition Two, WT%</u>	<u>Number of Dips into 27°C Tap Water to Clean Coated Lapshear</u>
0	50+
1	17
3	15
5	12
7	10
9	10

Example 5

An anaerobic sealant composition according to the present invention (Composition Three) was prepared with the following formulation:

Table 7

<u>Inventive Composition Three</u>	<u>Anaerobic Sealant, WT %</u>
Glycerol dimethacrylate	97.0
cumene hydroperoxide	<u>3.0</u>
Total:	100.0

Two drops, or about 0.4 grams, of the inventive composition was placed on a lapshear, as described in Example 1. A second lapshear was placed over the inventive composition, and the two lapshears were clamped together. A fixture test was then preformed at periodic time intervals. For the fixture test the two lapshears were unclamped at a particular time period. If the lapshears could move relative to one and the other, the inventive composition did not fully cure. The two lapshear would be reclamped until the next time interval. If the two unclamped lapshears could not moved relative to one and the other, then the inventive composition did fully cure. As illustrated below results in Table 8, the inventive composition anaerobically cured and proved to have particular efficacy as an impregnation composition with improved washability characteristics.

Table 8

<u>Inventive Composition Three</u>	<u>Room Temperature Anaerobic Curing</u>
1 hour after assembly	Not Cured
2 hours after assembly	Not Cured
3 hours after assembly	Not Cured
4 hours after assembly	Fully Cured

Example 6

- 5 An anaerobic sealant composition according to the present invention (Composition Four) was prepared with the following formulation:

Table 9

<u>Inventive Composition Four</u>	<u>Anaerobic Sealant, WT %</u>
Glycerol dimethacrylate	96.7
cumene hydroperoxide	3.0
Benzosulfimide (saccharin)	<u>0.3</u>
Total:	100.0

- 10 Two drops, or about 0.4 grams, of the inventive composition was used for a fixture test, as described in Example 5, with two lapshears. Saccharin proved to be an effective accelerator for aerobically curing the inventive composition. As illustrated below results in Table 10, the inventive composition anaerobically cured and proved to have particular efficacy as an impregnation composition with improved washability characteristics.

Table 10

<u>Inventive</u> <u>Composition Four</u> 1 hour after assembly	<u>Room</u> <u>Temperature</u> <u>Anaerobic Curing</u> Fully Cured
--	---

Example 7

An anaerobic sealant composition according to the present invention (Composition
5 Five) was prepared by adding 1 gram of copper octanoate to 100 grams of Inventive
Composition Four of Example 6. Two drops, or about 0.4 grams, of the inventive
composition was used for a fixture test, as described in Example 5, with two lapshears.
Copper octanoate proved to be an effective accelerator for aerobically curing the inventive
composition. As illustrated below results in Table 11, the inventive composition anaerobically
10 cured and proved to have particular efficacy as an impregnation composition with improved
washability characteristics.

Table 11

<u>Inventive</u> <u>Composition Five</u> 20 minutes after assembly	<u>Room</u> <u>Temperature</u> <u>Anaerobic Curing</u> Fully Cured
--	---

Example 8

An anaerobic sealant composition according to the present invention (Composition Six) was prepared with the following formulation:

5

Table 12

<u>Inventive Composition Six</u>	<u>Anaerobic Sealant, WT %</u>
Glycerol dimethacrylate	77.0
Lauryl methacrylate	10.0
Hydroxypropyl methacrylate	10.0
cumene hydroperoxide	<u>3.0</u>
Total:	100.0

Two drops, or about 0.4 grams, of the inventive composition was used for a fixture test, as described in Example 5, with two lapshears. As illustrated below results in Table 13, the inventive composition anaerobically cured and proved to have particular efficacy as an impregnation composition with improved washability characteristics.

10

Table 13

<u>Inventive Composition Six</u>	<u>Room Temperature Anaerobic Curing</u>
1 hour after assembly	Not Cured
2 hours after assembly	Not Cured
3 hours after assembly	Not Cured
4 hours after assembly	Not Cured
5 hours after assembly	Not Cured
6 hours after assembly	Not Cured
7 hours after assembly	Partially Cured

Example 9

An heat curable sealant composition according to the present invention (Composition Seven) was prepared with the following formulation:

5

Table 14

Inventive <u>Composition Seven</u>	Heat Curable Sealant, <u>WT %</u>
Glycerol dimethacrylate	97.0
cumene hydroperoxide	<u>3.0</u>
Total:	100.0

Two drops, or about 0.4 grams, of the inventive composition was placed on a lapshear, as described in Example 1. A second lapshear was placed over the inventive composition, and the two lapshears were clamped together. The assembly was placed in an oven and maintained at 121°C. A fixture test, as described in Example 5, was then preformed after one hour. As illustrated below results in Table 15, the inventive composition thermally cured at 121°C and proved to have particular efficacy as an impregnation composition with improved washability characteristics.

10

15

Table 15

Inventive <u>Composition Seven</u> 1 hour after assembly	121 °C <u>Heat Curing</u> Fully Cured
--	---

Example 10

A heat curable sealant composition according to the present invention (Composition

Eight) was prepared with the following formulation:

Table 16

Inventive <u>Composition Eight</u>	Heat Curable Sealant, <u>WT %</u>
Glycerol dimethacrylate	96.7
cumene hydroperoxide	3.0
Benzosulfimide (saccharin)	<u>0.3</u>
Total:	100.0

Two drops, or about 0.4 grams, of the inventive composition was used for a fixture test, as described in Example 9, with two lapshears. As illustrated below results in Table 17, the inventive composition thermally cured at 121°C and proved to have particular efficacy as an impregnation composition with improved washability characteristics.

Table 17

Inventive <u>Composition Eight</u> 1 hour after assembly	121°C <u>Heat Curing</u> Fully Cured
--	--

Example 11

A heat curable sealant composition according to the present invention (Composition Nine) was prepared with the following formulation:

Table 18

<u>Inventive Composition Nine</u>	<u>Heat Curable Sealant, WT %</u>
Glycerol dimethacrylate	77.0
Lauryl methacrylate	10.0
Hydroxpropyl methacrylate	10.0
cumene hydroperoxide	<u>3.0</u>
Total:	100.0

Two drops, or about 0.4 grams, of the inventive composition was used for a fixture test, as described in Example 9, with two lapshears. As illustrated below results in Table 19,
5 the inventive composition thermally cured at 121°C and proved to have particular efficacy as an impregnation composition with improved washability characteristics.

Table 19

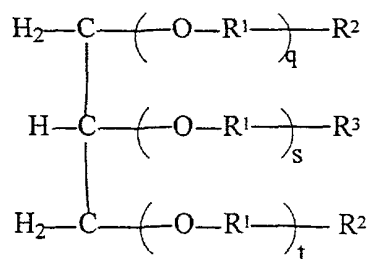
<u>Inventive Composition Nine</u> 1 hour after assembly	<u>121°C Heat Curing Fully Cured</u>
--	--

10 The invention being thus described, it will be clear to those persons of skill in the art that the same may be varied in many ways. Such variations are not to be regarded as a departure from the spirit and scope of the invention and all such modifications are intended to be included within the scope of the claims.

WHAT IS CLAIMED IS:

1. A free-radical curable composition which is washable and self-emulsifiable upon mixing with water comprising:

5 (a) a curable glycerol composition having the formula:



10 wherein R¹ is a substituted or unsubstituted C₁ to C₅ alkyl or combinations thereof; R² and R³ are independently selected from the group consisting of hydroxyl, (meth)acrylate and combinations thereof; q, s and t are independently from about 0 to about 35; provided that at least one of said R² is said (meth)acrylate; and

(b) a free radical initiator to initiate cure of said composition.

15 2. The composition of claim 1 wherein said free radical initiator includes a heat-curing initiator to produce free radicals by thermal decomposition to cure said sealant.

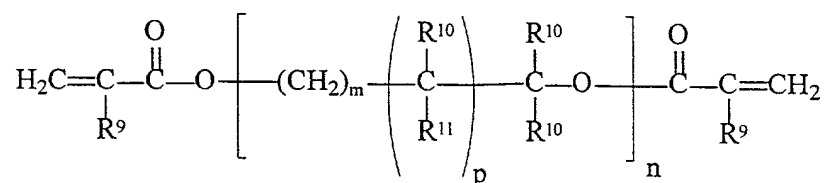
20 3. The composition of claim 2 wherein the heat-curing initiator is selected from the group consisting of a peroxide, a hydroperoxide, a perester, an azonitrile and combinations thereof.

4. The composition of claim 1 wherein said free radical initiator includes a anaerobic-curing initiator to produce free radicals upon the exclusion of oxygen to cure said sealant.

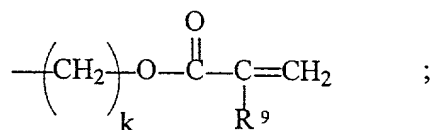
5. The composition of claim 4 wherein said anaerobic-curing initiator is a peroxy initiator selected from the group consisting of hydroperoxides, peroxides, peresters and combinations thereof.

6. The composition of claim 4 wherein said anaerobic-curing initiator includes an anaerobic accelerator selected from the group consisting of tributyl amine, benzoic sulfimide, formamide, copper octanoate and combinations thereof.

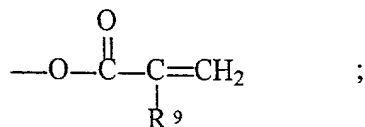
7. The composition of claim 1 further including a poly(meth)acrylate ester having the formula:



wherein R¹⁰ represents a radical selected from the group consisting of hydrogen, lower alkyl of from 1 to about 4 carbon atoms, hydroxyalkyl of from 1 to about 4 carbon atoms and



R⁹ is a radical selected from the group consisting of hydrogen, halogen, and lower alkyl of from 1 to about 4 carbon atoms; R¹¹ is a radical selected from the group consisting of hydrogen, hydroxyl and



m is 0 to about 12, n is equal to at least 1, k is 1 to about 4 and p is 0 or 1.

8. The composition of claim 1 further including a monofunctional acrylate ester, said monofunctional acrylate ester being selected from the group consisting of lauryl

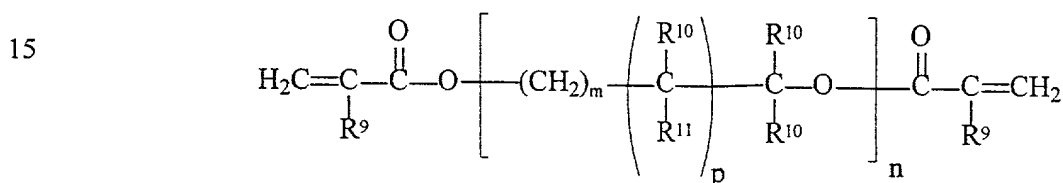
methacrylate, cyclohexylmethacrylate, tetrahydrofurfuryl methacrylate, hydroxyethyl acrylate, hydroxypropyl methacrylate, t-butylaminoethyl methacrylate, cyanoethylacrylate, chloroethylmethacrylate and combinations thereof.

5 9. The composition of claim 1 further including an ionic surfactant, an anionic surfactant and combinations thereof.

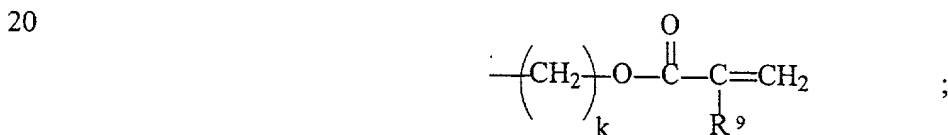
10 10. The composition of claim 1 wherein R¹ is ethyl, propyl or a combination thereof.

10 11. A free-radical curable composition which is washable and self-emulsifiable upon mixing with water comprising:

(a) a curable poly(meth)acrylate ester having the formula:

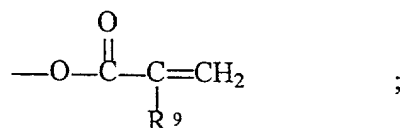


wherein R¹⁰ represents a radical selected from the group consisting of hydrogen, lower alkyl of from 1 to about 4 carbon atoms, hydroxyalkyl of from 1 to about 4 carbon atoms and



R⁹ is a radical selected from the group consisting of hydrogen, halogen, and lower alkyl of from 1 to about 4 carbon atoms; R¹¹ is a radical selected from the group consisting of

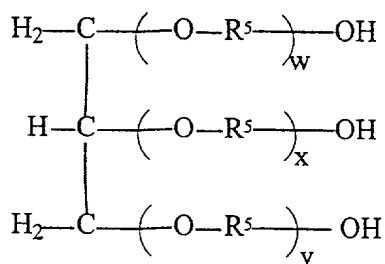
25 hydrogen, hydroxyl and



m is 0 to about 12, n is equal to at least 1, k is 1 to about 4 and p is 0 or 1;

(b) a washing agent for emulsifying said curable poly(meth)acrylate ester, said washing agent is selected from the group consisting of a glycerol composition, a (meth)acrylate glycerol composition and combinations thereof; wherein

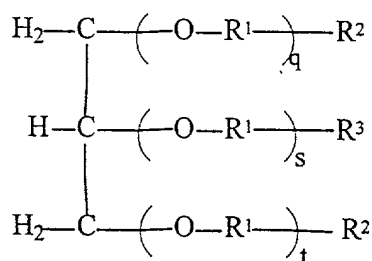
(i) said glycerol composition having the formula:



5

wherein R^5 is a C_1 to C_5 substituted or unsubstituted alkyl or a combination thereof; w , x and y are independently from 0 to about 35; and

(ii) said (meth)acrylate glycerol composition having the formula:



wherein R^1 is a substituted or unsubstituted C_1 to C_5 alkyl or combinations thereof; R^2 and R^3 are independently selected from the group consisting of hydroxyl, (meth)acrylate and combinations thereof; q , s and t are independently from about 0 to about 35; provided that at least one of said R^2 is said (meth)acrylate; and

10

(c) a free radical initiator for producing free radicals to initiate cure said

15

composition.

12. The composition of claim 11 wherein said free radical initiator includes a heat-curing initiator to produce free radicals by thermal decomposition to cure said composition.

13. The composition of claim 12 wherein the heat-curing initiator is selected from the group consisting of a peroxide, a hydroperoxide, a perester, an azonitrile and combinations thereof.

5 14. The composition of claim 11 wherein said free radical initiator includes a anaerobic-curing initiator to produce free radicals upon the exclusion of oxygen to cure said sealant

10 15. The composition of claim 14 wherein said anaerobic-curing initiator is a peroxy initiator selected from the group consisting of hydroperoxides, peroxides, peresters and combinations thereof.

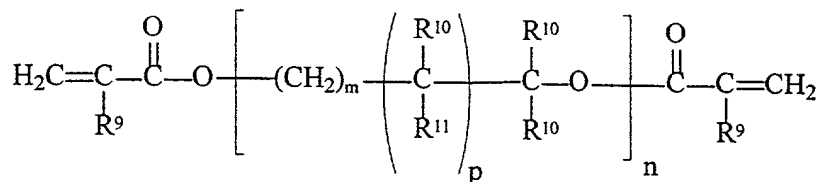
15 16. The composition of claim 14 wherein said anaerobic-curing initiator includes an anaerobic accelerator selected from the group consisting of tributyl amine, benzoic sulfimide, formamide, copper octanoate and combinations thereof.

20 17. The composition of claim 11 further including a monofunctional acrylate ester, said monofunctional acrylate ester being selected from the group consisting of lauryl methacrylate, cyclohexylmethacrylate, tetrahydrofurfuryl methacrylate, hydroxyethyl acrylate, hydroxypropyl methacrylate, t-butylaminoethyl methacrylate, cyanoethylacrylate, chloroethylmethacrylate and combinations thereof.

18. The composition of claim 11 further including an ionic surfactant, an anionic surfactant and combinations thereof.

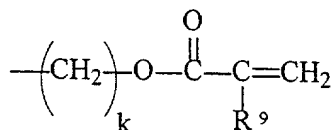
19. A method for washing uncured anaerobic or heat curing sealant from a surface from an article comprising:

(a) impregnating said article with a curable poly(meth)acrylate ester having the formula:

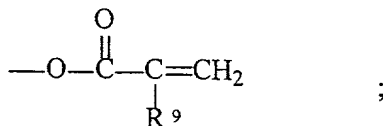


5

wherein R^{10} represents a radical selected from the group consisting of hydrogen, lower alkyl of from 1 to about 4 carbon atoms, hydroxyalkyl of from 1 to about 4 carbon atoms and



10 R^9 is a radical selected from the group consisting of hydrogen, halogen, and lower alkyl of from 1 to about 4 carbon atoms; R^{11} is a radical selected from the group consisting of hydrogen, hydroxyl and

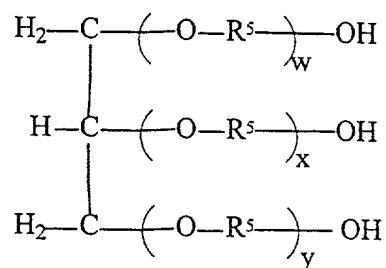


15 m is 0 to about 12, n is equal to at least 1, k is 1 to about 4, and p is 0 or 1;

(b) adding a washing agent for emulsifying said curable poly(meth)acrylate ester into a wash tank containing an aqueous solution, said washing agent is selected from the group consisting of a glycerol composition, a (meth)acrylate glycerol composition and combinations thereof; wherein

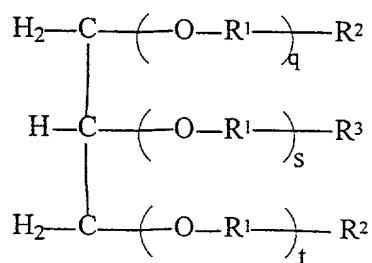
20

(i) said glycerol composition having the formula:



wherein R⁵ is a substituted or unsubstituted C₁ to C₅ alkyl or a combination thereof; w, x and y are independently from about 0 to about 35 and

(ii) said (meth)acrylate glycerol composition having the formula:

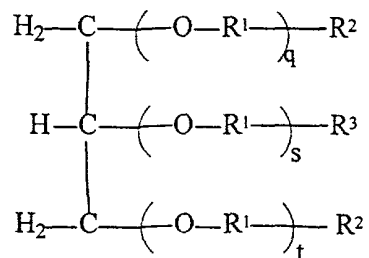


wherein R¹ is a substituted or unsubstituted C₁ to C₅ alkyl or combinations thereof; R² and R³ are independently selected from the group consisting of hydroxyl, (meth)acrylate and combinations thereof; q, s and t are independently from about 0 to about 35; provided that at least one of said R² is said (meth)acrylate; and

(c) washing said curable poly(meth)acrylate ester from the surface of said article in said wash tank containing said aqueous solution and said washing agent.

20. A method of anaerobically or thermally sealing a porous article comprising:

(a) selecting a curable glycerol composition having the formula:



wherein R¹ is a substituted or unsubstituted C₁ to C₅ alkyl or combinations thereof; R² and R³

5 are independently selected from the group consisting of hydroxyl, (meth)acrylate and combinations thereof; q, s and t are independently from about 0 to about 35; provided that at least one of said R² is said (meth)acrylate; and

10 (b) selecting a free radical initiation to initiate curing of said curable glycerol;

(c) impregnating pores of said article with said curable glycerol and said initiator, and

15 (d) washing said curable glycerol from a surface of said article in a wash tank containing an aqueous solution.

COMBINED DECLARATION AND POWER OF ATTORNEY

(ORIGINAL, DESIGN, NATIONAL STAGE OF PCT, SUPPLEMENTAL, DIVISIONAL, CONTINUATION OR CIP)

As a below named inventor, I hereby declare that:

TYPE OF DECLARATION

This declaration is of the following type:

- ☐ original
- ☐ design
- ☐ supplemental
- ☒ national stage of PCT
- ☐ divisional
- ☐ continuation
- ☐ continuation-in-part (CIP)

INVENTORSHIP IDENTIFICATION

My residence, post office address and citizenship are as stated below next to my name, I believe I am the original, first and sole inventor (*if only one name is listed below*) or an original, first and joint inventor (*if plural names are listed below*) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

TITLE OF INVENTION

Washable Impregnation Compositions

SPECIFICATION IDENTIFICATION

the specification of which:

- (a) ☐ is attached hereto.
- (b) ☐ was filed on _____ as ☐ Application No. _____ or ☐ Express Mail Mailing Label No. _____, as Application No. not yet known, and was amended on _____ (*if applicable*).
- (c) ☒ was described and claimed in PCT International Application No. PCT/US00/20076 filed on 21 July 2000 and as amended under PCT Article 19 on _____ (*if any*).

ACKNOWLEDGMENT OF REVIEW OF PAPERS AND DUTY OF CANDOR

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations § 1.56(a).

**COMBINED DECLARATION
AND POWER OF ATTORNEY**

Attorney Docket No.: LC-377/PCT/US

PRIORITY CLAIM

I hereby claim foreign priority benefits under Title 35, United States Code, § 119(a)-(d) of any foreign application(s) for patent or inventor's certificate, § 119(e) of any United States provisional application, or § 365(b) of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

(d) ☐ no such applications have been filed.

(e) ☒ such applications have been filed as follows:

**EARLIEST FOREIGN APPLICATION(S), IF ANY, FILED WITHIN 12 MONTHS
(6 MONTHS FOR DESIGN) PRIOR TO THIS U.S. APPLICATION**

COUNTRY	APPLICATION NUMBER	DATE OF FILING (day,month,year)	PRIORITY CLAIMED UNDER 35 USC 119
US	60/144,896	27 Sept 2000	<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No
			<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No
			<input type="checkbox"/> Yes <input type="checkbox"/> No

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s), or § 365 (c) of any PCT International application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of Title 35, United States Code, § 112.

(f) ☐ no such applications have been filed.

(g) ☒ such applications have been filed as follows:

APPLICATION NUMBER	DATE OF FILING (day, month, year)	PRIORITY CLAIMED UNDER 35 USC 120
PCT/US00/20076	21 July 2000	<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No
		<input type="checkbox"/> Yes <input type="checkbox"/> No

POWER OF ATTORNEY

As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith:

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COMBINED DECLARATION
AND POWER OF ATTORNEY

Attorney Docket No.: LC-377/PCT/US

DECLARATION

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

SIGNATURES

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Inventor's signature: *Frederick F. Newberth III*

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Full name of second inventor: Peter J. Chupas

Inventor's signature: _____

Date: _____ Country of Citizenship: United States

Residence: _____

Post Office Address: 4 Rosita Lane, Port Jefferson, NY 11777

100438888 80887001

COMBINED DECLARATION AND POWER OF ATTORNEY*(ORIGINAL, DESIGN, NATIONAL STAGE OF PCT, SUPPLEMENTAL, DIVISIONAL, CONTINUATION OR CIP)*

As a below named inventor, I hereby declare that:

TYPE OF DECLARATION

This declaration is of the following type:

- ☐ original
- ☐ design
- ☐ supplemental
- ☒ national stage of PCT
- ☐ divisional
- ☐ continuation
- ☐ continuation-in-part (CIP)

INVENTORSHIP IDENTIFICATION

My residence, post office address and citizenship are as stated below next to my name. I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled.

TITLE OF INVENTION

Washable Impregnation Compositions

SPECIFICATION IDENTIFICATION

the specification of which:

- (a) ☐ is attached hereto.
- (b) ☐ was filed on _____ as Application No. _____ or Express Mail Mailing Label No. _____, as Application No. not yet known, and was amended on _____ (if applicable).
- (c) ☒ was described and claimed in PCT International Application No. PCT/US00/20076 filed on 21 July 2000 and as amended under PCT Article 19 on _____ (if any).

ACKNOWLEDGMENT OF REVIEW OF PAPERS AND DUTY OF CANDOR

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations § 1.56(a).

COMBINED DECLARATION
AND POWER OF ATTORNEY

Attorney Docket No.: 1.C-377/PCT/US

PRIORITY CLAIM

I hereby claim foreign priority benefits under Title 35, United States Code, § 119(a)-(d) of any foreign application(s) for patent or inventor's certificate, § 119(e) of any United States provisional application, or § 365(b) of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

(d) ☐ no such applications have been filed.

(c) ☒ such applications have been filed as follows:

EARLIEST FOREIGN APPLICATION(S), IF ANY, FILED WITHIN 12 MONTHS
(6 MONTHS FOR DESIGN) PRIOR TO THIS U.S. APPLICATION

COUNTRY	APPLICATION NUMBER	DATE OF FILING (day, month, year)	PRIORITY CLAIMED UNDER 35 USC 119
US	60/144,896	27 Sept 2000	<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No
			<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No
			<input type="checkbox"/> Yes <input type="checkbox"/> No

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s), or § 365 (c) of any PCT International application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of Title 35, United States Code, § 112.

(f) ☐ no such applications have been filed.

(g) ☒ such applications have been filed as follows:

APPLICATION NUMBER	DATE OF FILING (day, month, year)	PRIORITY CLAIMED UNDER 35 USC 120
PCT/US00/20076	21 July 2000	<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No
		<input type="checkbox"/> Yes <input type="checkbox"/> No

POWER OF ATTORNEY

As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith.

Steven C. Bauman
Reg. No. 33,832
LOCTITE CORPORATION
1001 Trout Brook Crossing
Rocky Hill, Connecticut 06067 USA

SEND CORRESPONDENCE TO:	DIRECT TELEPHONE CALLS TO: (name and telephone number)
LOCTITE CORPORATION Legal Department 1001 Trout Brook Crossing Rocky Hill, Connecticut 06067	Steven C. Bauman (860) 571-5001

COMBINED DECLARATION AND POWER OF ATTORNEY

Attorney Docket No.: LC-377/PCT/US

DECLARATION

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

SIGNATURES

Full name of sole or first inventor: Frederick F Newberth III

Inventor's signature: _____

Date: _____ Country of Citizenship: United States

Residence: 12 Greenhurst Road, West Hartford, CT 06107

Post Office Address: _____

Full name of second inventor. Peter J. Chupas

Inventor's signature: Pete J. Chappin

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